Prediction of Metal Partitioning between Minerals and Aqueous Solutions: A Modeling Tool for Selecting Effective Containment Materials

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Trace metal partitioning between authigenic minerals and aqueous solutions is an important chemical process for waste containment. In this paper, we have developed a linear free energy correlation model that correlates metal partition coefficients with metal cation properties:

$$-2.303RT\log K_d = a^*_{M_HX}(\Delta G^0_{n,M^{Z+}} - \Delta G^0_{n,M_H^{Z+}}) + \beta^*_{M_HX}(r_{M^{Z+}} - r_{M_H^{Z+}}) - (\Delta G^0_{f,M^{Z+}} - \Delta G^0_{f,M_H^{Z+}})$$

where $a^*_{M_HX}$ and $\beta^*_{M_HX}$ are constants and can be determined by a regression analysis. Host minerals from an isostructural family have the same linear free energy relationship, as long as the relationship is expressed as a function of the differences in cation properties between substituent and host metals. We have applied our model to both isovalent and non-isovalent metal partitioning in carbonate minerals. The model closely fits experimental data, demonstrating the robustness of the proposed linear free energy relationship. Using the model, we have predicted the partition coefficients of divalent and trivalent metals between various carbonate minerals and aqueous solutions. Magnesite is predicted to have the largest partition coefficients among the carbonate minerals with a calcite structure and therefore can be a good scavenger for toxic metals. The implication of this prediction to the Waste Isolation Pilot Plant is discussed. The model developed in this paper provides a useful tool for predicting unknown partition coefficients and therefore help select effective containment materials.